

Reply to Comment on Circular Dichroism in the Angle-Resolved Photoemission Spectrum of the High-Temperature $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ Superconductor, <http://arxiv.org/abs/1004.1648>

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Our purpose in Ref. 1 was to carry out a realistic first-principles angle-resolved photointensity (ARPES) computation to assess the importance of geometric effects in inducing dichroism in the cuprates, and not so much to focus on the results of Ref. 2. Using the known low- and high-temperature (LT and HT) bulk crystal structures of $\text{Bi}_2\text{212}$, we showed that the orthorhombic distortion in the Bi-O plane in going from the HT to the LT structure yields an increase in the dichroic signal of 1.5 to 3.5%, depending on photon energy, which is comparable to the dichroic effect of 3% reported in Ref. 2. Moreover, our computed dichroic signal of 6% at 49.7 eV photon energy for the HT structure is comparable to the 5% effect reported on single crystals [3]. Thus, we clearly established the sensitivity of dichroism to structural details and the viability of the geometric mechanism in explaining existing measurements, and demonstrated that the detection of time reversal symmetry breaking via ARPES will be complicated by the masking effects of lattice distortions.

The structure of thin films of Ref. 2, especially how this structure varies with temperature/doping, is unknown. That the films were heavily twinned is revealed by the authors for the first time in their present Comment, but how the population of the two twins varies with doping or temperature is not known. Since the films display weak superstructure, one may speculate that these films are structurally closer to Pb-doped rather than non-Pb-doped crystals. Unfortunately, however, experiments on Pb-doped crystals [3] found a zero dichroic signal at low- as well as high-T, suggesting that the observed dichroism in Ref. 2 is peculiar to films. To date, the thin-film measurement of Ref. 2 is the only ARPES experiment in the literature reporting a non-zero dichroic effect at low-T.

We emphasize that a 1% dichroic effect can be induced by an in-plane movement of O-atoms in the BiO layer by only 0.03 Å, which is well below the accuracy with which lateral positions of surface atoms can be determined currently via surface sensitive probes. Given the sensitivity of dichroism to structural details it will be prudent to keep the following further points in mind in interpreting dichroic effects and their temperature/doping dependencies: (1) ARPES is a surface-sensitive probe, and the surface structure of thin-films or single crystals will in general be different from bulk. We employed an ideal

Bi-O terminated semi-infinite crystal in our computations because the detailed surface structure is unknown (x-ray scattering is not sensitive to surface structure); (2) Surface structure will likely be more sensitive than bulk to temperature; (3) Ref. 2 reports sample to sample variation in dichroism, which they ascribe to changes in domain structure. If so, such domain variations may become exaggerated in the surface region. Our simulations show that 1% dichroic effect can result from only a 4% change in the population of one twin; (4) Additional complications involve effects of disorder and layer-to-layer variations in structure in the surface region.

We would expect the computed momentum and energy dependencies of the dichroic signal to be even more sensitive to structural details than the size of the dichroic signal. Therefore, the detailed comparisons of the sort alluded to in the Comment between our computations on an ideal Bi-O terminated bulk structure and experiments on thin films, where the computations and measurements refer to different structures, are not of much relevance.

Finally, we note that structural issues with the study of Ref. 2 have been raised earlier in the comments contained in Refs. 4 and 5. We refer to those comments and responses for details of these and other issues raised previously in the literature on the study of Ref. 2.

We conclude that arguments of Norman et al. in their present Comment do not provide a significant basis for their claim that the geometric mechanism for explaining the observations reported in Ref. 2 is not viable. More generally, our study [1] highlights the importance of assessing structural issues before invoking exotic mechanisms for explaining unusual spectroscopic observations, especially in complex materials.

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